

CLAIMS

What is claimed is:

1. A gas desulfurization assembly for recovering H_2S from a waste gas stream, said assembly comprising:

a short contact time catalytic reactor having

a H_2S -containing gas injection inlet,

an O_2 -containing gas injection inlet,

a gas mixing zone, and

a reaction zone comprising at least one catalyst device having activity for catalyzing the partial oxidation of H_2S to elemental sulfur and water under reaction promoting conditions;

a cooling zone; and

a sulfur condenser in fluid communication with said cooling zone and having a liquid sulfur outlet and a desulfurized gas outlet, said injection inlets being in fluid communication with said gas mixing zone, said mixing zone being in fluid communication with said reaction zone, and said reaction zone being in fluid communication with said cooling zone, and said cooling zone being in fluid communication with said sulfur condenser.

2. The assembly of claim 1 further comprising at least one tail gas treatment unit in fluid communication with said sulfur condenser.

3. The assembly of claim 1 further comprising a heater disposed between said desulfurized gas outlet and said tail gas treatment unit.

4. The assembly of claim 1 wherein at least the reaction zone of said reactor is capable of withstanding temperatures of at least about $700^{\circ}C$.

5. The assembly of claim 1 wherein said reactor comprises a thermal shield between said mixing zone and said reaction zone.

6. The assembly of claim 1 wherein said cooling zone comprises a plurality of thermally conductive tubes.

7. The assembly of claim 1 wherein said cooling zone further comprises at least one thermal insulator between said reaction zone and said thermally conductive tubes.

8. The assembly of claim 5 wherein said at least one thermal insulator comprises a plurality of refractory ferrules each of which is attached to a thermally conductive tube.

9. The assembly of claim 1 wherein said cooling zone comprises a heat exchanger.

10. A waste gas desulfurization process comprising:

providing a H_2S -containing waste gas stream;

providing a O_2 -containing stream;

in a millisecond contact time reactor having a gas mixing zone, a reaction zone, and a cooling zone, mixing together said H_2S -containing gas stream and said O_2 -containing gas stream in said mixing zone to form a reactant gas mixture, said reaction zone containing a catalyst device having activity for catalyzing the partial oxidation of H_2S to elemental sulfur and water;

maintaining the temperature of said reaction zone above 300°C ;

passing said reactant gas mixture over said catalyst device such that the contact time between said catalyst device and a portion of said reactant gas mixture that contacts said catalyst device is sufficiently brief to allow the reaction $\text{H}_2\text{S} + 1/2 \text{O}_2 \rightarrow 1/x \text{S}_x + \text{H}_2\text{O}$ ($x = 2, 6$ or 8) to occur, and a product gas stream is formed comprising gaseous elemental sulfur and water;

passing said product gas stream into said cooling zone and cooling said product stream to a temperature above the dewpoint of sulfur, to provide a partially cooled product stream;

passing said partially cooled product stream into a sulfur condenser and further cooling said partially cooled product stream to the dewpoint temperature of elemental sulfur, or lower.

11. The process of claim 10 wherein at least the reaction zone of said reactor is capable of withstanding temperatures of at least $1,500^\circ\text{C}$, said H_2S -containing gas stream and said O_2 -

containing gas stream are combined in said mixing zone to form a reactant gas mixture having a molar ratio of H_2S to O_2 of about 2:1 or less, and the temperature of said reaction zone is 700-1,500°C.

12. The process of claim 10 comprising preheating said H_2S and/or O_2 stream up to about 200°C before contacting said catalyst device.

13. The process of claim 10 wherein said contact time is no more than about 200 milliseconds.

14. The process of claim 10 comprising recovering a product comprising elemental sulfur and, optionally, venting a substantially desulfurized residual gas.

15. The process of claim 10 comprising providing a H_2S -containing gas containing at least about 1 vol.% H_2S .

16. The process of claim 10 wherein said O_2 -containing gas is chosen from the group consisting of purified O_2 , air, and O_2 enriched air.

17. The process of claim 10 further comprising maintaining the temperature of said reaction zone between about 850°C - 1,300°C.

18. The process of claim 17 further comprising regulating reactor inlet temperatures, regulating H_2S concentration in the reactant gas mixture, and/or applying heat to said catalyst device such that the temperature of said catalyst device is maintained at a desired temperature between said 850-1,300°C.

19. The process of claim 17 further comprising maintaining autothermal reaction promoting conditions.

20. The process of claim 10 wherein said catalyst device comprises at least one metal chosen from the group consisting of platinum, rhodium, ruthenium, iridium, nickel, palladium, iron, cobalt, rhenium, rubidium, vanadium, bismuth and antimony.

21. The process of claim 20 wherein said catalyst device comprises at least one metal chosen from the group consisting of platinum, rhodium, ruthenium, iridium, nickel, palladium, iron, cobalt, rhenium and rubidium.

22. The process of claim 21 wherein said catalyst device comprises at least one metal chosen from the group consisting of platinum, rhodium, ruthenium, nickel, palladium and iridium

23. The process of claim 22 wherein said catalyst device comprises platinum, rhodium or a mixture thereof.

24. The process of claim 20 wherein said catalyst device comprises vanadium, bismuth or antimony.

25. The process of claim 20 wherein said catalyst device comprises at least one lanthanide element chosen from the group consisting of La, Ce, Pr, Nd, Pm, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb and Lu.

26. The process of claim 25 wherein at least one said lanthanide element is samarium, ytterbium or praseodymium.

27. The process of claim 10 wherein said catalyst device comprises at least one structure chosen from the group consisting of gauzes, monoliths and a plurality of divided units.

28. The process of claim 27 wherein said divided units comprise particles, granules, beads, pills, pellets, cylinders, trilobes, extrudates or spheres.

29. The process of claim 27 wherein said structure comprises a refractory support.

30. The process of claim 29 wherein said catalyst refractory support comprises zirconia or alumina.

31. The process of claim 10 wherein said catalyst device comprises platinum and a lanthanide metal oxide chosen from the group consisting of cerium oxide and lanthanum oxide.

32. The process of claim 31 wherein said catalyst device comprises rhodium and samarium oxide.

33. The process of claim 10 wherein said catalyst device comprises a platinum-rhodium alloy disposed on a lanthanide oxide coated refractory support.

34. The process of claim 33 wherein said catalyst device comprises a samarium oxide coated refractory support.

35. The process of claim 10 wherein said catalyst device comprises at least one carbided metal.

36. The process of claim 35 wherein said carbided metal comprises platinum and rhodium.

37. The process of claim 10 comprising operating said reactor at a space velocity of at least about 20,000 hr⁻¹.

38. The process of claim 10 comprising operating said reactor at superatmospheric pressure.

39. A method of preparing a catalyst device that is active for catalyzing the direct partial oxidation of hydrogen sulfide to elemental sulfur and water, the process comprising:

contacting a precursor device at a temperature greater than 300°C, with a carbiding gas comprising a light hydrocarbon, said precursor device containing at least one reduced metal chosen from the group consisting of platinum, rhodium, ruthenium, iridium, nickel, palladium,

iron, cobalt, rhenium, rubidium, vanadium, bismuth and antimony, such that a carbided catalyst device is produced.

40. The method of claim 39 wherein said carbiding gas comprises a mixture of said light hydrocarbon and O₂ in a carbon:oxygen molar ratio of 2:1.

41. The method of claim 39 wherein said carbiding gas flows over said catalyst precursor device.

42. The method of claim 41 wherein said carbiding gas flows over said precursor catalyst device at such space velocity that each portion of said carbiding gas that contacts said device resides on said device no more than 200 milliseconds.

43. The method of claim 39 wherein said device comprises at least one layer of gauze, at least one monolith or at least one layer of a plurality of divided units.

44. The method of claim 43 wherein each said divided unit is less than 10 millimeters in its longest dimension.

45. The method of claim 39 wherein said light hydrocarbon comprises methane, ethane, propane or butane.

46. The method of claim 39 wherein said metal comprises a platinum-rhodium alloy.

47. The method of claim 39 wherein said contacting is carried out in the short contact time reactor of the gas desulfurization assembly of claim 1.

48. The method of claim 39 wherein said contacting comprises a temperature of at least 700°C.

49. The method of claim 39 wherein said contacting comprises a temperature of at least 850°C.

50. The method of claim 39 wherein said contacting comprises a temperature up to about 1,500°C.

51. The method of claim 39 wherein said contacting includes initiating the catalytic partial oxidation of said light hydrocarbon to form carbon monoxide and hydrogen.

52. The method of claim 39 comprising ensuring that a H₂S-containing gas does not contact said catalyst precursor device before said catalyst precursor device is contacted with said light hydrocarbon.

53. The process of claim 10 wherein said catalyst device comprises a carbided catalyst device prepared according to the method of claim 39.

54. The process of claim 53 comprising, in the same reactor, stopping the flow of said carbiding gas and beginning flow of H₂S containing gas over said carbided catalyst device at a temperature sufficient to initiate H₂S catalytic partial oxidation to form elemental sulfur and water.